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OPTICAL, ELECTRICAL AND MICROSTRUCTURAL PROPERTIES OF TIN DOPED INDIUM OXIDE FILMS MADE FROM SINTERED NANOPARTICLES

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ABSTRACT

Thin transparent and electrically conductive films of tin doped indium oxide (ITO) were made by sintering of nanoparticle dispersions. The resistivity decreased to $1 \cdot 10^{-2}$ Ωcm upon treatment at 800°C, while the luminous transmittance remained high. The property evolution was connected with sintering and densification as studied by Scanning Electron Microscopy, X-ray Diffraction, X-ray Fluorescence and Elastic Recoil Detection Analysis.

INTRODUCTION

Films of tin doped indium oxide (ITO) are able to combine high visible transparency with good electrical conductivity. They are widely used in modern technology such as for flat panel displays, solar cells, switchable and low-emissivity windows, as well as for electromagnetic shielding [1]. The most prevalent production methods for ITO films are sputtering and evaporation.

Many applications, including flat panel displays, require patterned ITO layers. The pattering is commonly achieved by etching which, however, is a time consuming and environmentally less favourable method. An attractive alternative would be to print the ITO structure directly onto a substrate.

The purpose of this work is to investigate the microstructural, optical, and electrical properties of thin films made from an ITO nanoparticle dispersion. The ultimate goal is to develop a technique allowing thin lines of ITO to be applied by printing.

FILM PREPARATION AND STRUCTURE

ITO particles were produced by a wet-chemical method. The average primary particle diameter and crystallite sizes were both approximately 16 nm. The tin to indium atomic ratio was 5%. 1.1 μm thick films were prepared by spin-coating a dispersion of the particles onto Corning 1737 glass and Al_2O_3 substrates. The thickness was measured by a mechanical stylus Dektak 3030 profilometer. Post deposition sintering was carried out in the $300°C \leq T_s \leq 800°C$ range for 2 hours. The temperature was ramped by 10°C per minute.

The crystalline structure was investigated by X-ray Diffraction using a Siemens D5000 unit operating with CuK$_\alpha$ radiation. The diffractograms were consistent with an In_2O_3 structure (Powder Diffraction File 06-0416). Crystallite sizes were evaluated from the (222) peak of ITO by Scherrer's method [2]. For all samples only a minor shift in the peak location, as compared to In_2O_3 was noticed. Hence, the enlargement of the lattice parameter due to tin atom incorporation is small.

Microstructures were studied with Scanning Electron Microscopy (SEM), using a Leo 1550 instrument. Figure 1 gives a high-resolution image of agglomerated ITO nanoparticles treated at 500°C. It is obvious that the particles are loosely packed, i.e. that the relative density γ is much less than unity. Quantitative measurements using Elastic Recoil Detection Analysis (see e.g. [3]) and X-ray Fluorescence [4] indicated that the relative density was approximately 30% for samples sintered at 300 to 500°C.

Figure 2 presents SEM micrographs for samples treated at 400, 650, and 800°C. The corresponding average particle sizes determined from these pictures are 16, 19, and 34 nm. However, at 650°C when the particles have started growing the crystallite size still remains at 16 nm. At 800°C the grain and crystallite sizes are again equal.

It is obvious that a fully dense, sintered structure has not yet been reached at 800°C. According to conventional sintering theory [5], the grain growth severely slows down the sintering rate.
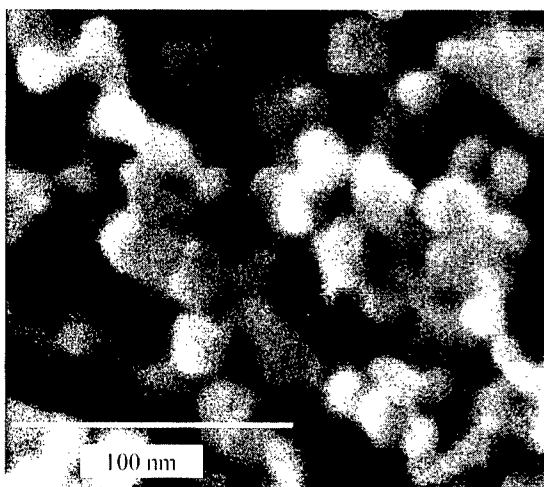

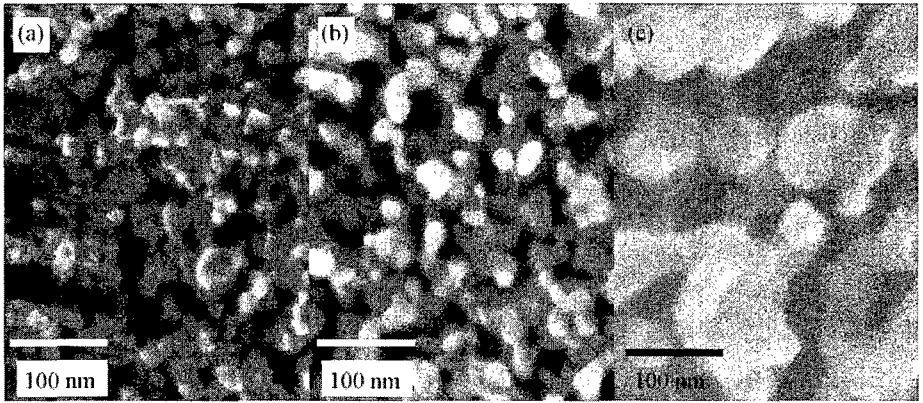**Figure 1**: High-resolution SEM image of nanoparticle-based ITO film, heat treated at 500°C.

Figure 2: SEM images of nanoparticle-based ITO films sintered at 400°C (a), 650°C (b) and 800°C (c).

OPTICAL AND ELECTRICAL PROPERTIES

Spectral normal transmittance (T) and near-normal reflectance (R) were measured in the $300 \leq \lambda \leq 2500$ nm wavelength range on a Perkin Elmer Lambda 9 spectrophotometer equipped by an integrating sphere. The resistivity (ρ) was calculated from resistance values measured by a square-probe. The data were taken on samples that had aged in room temperature and ambient atmosphere for several months. This generally means that the value of the resistivity has increased by a factor three, as compared to newly sintered samples. No effect of the specific substrate material was noticed.

Figure 3 illustrates $T(\lambda)$ and $R(\lambda)$ for five different sintering temperatures. It is found that $T(\lambda)$ is high across the $400 < \lambda < 700$ nm interval, pertinent to luminous radiation, and significant changes occur only at 800°C. Values of the luminous transmittance T_{lum}, determined by integrating over the sensitivity of the eye, are reported in Figure 4. Clearly, T_{lum} exceeds 90% except when the sintering temperature is 800°C.

The magnitude of the resistivity drops monotonically, as the sintering temperature is increased, as apparent from Figure 4, and reaches $10^{-2}\Omega$cm at 800°C.
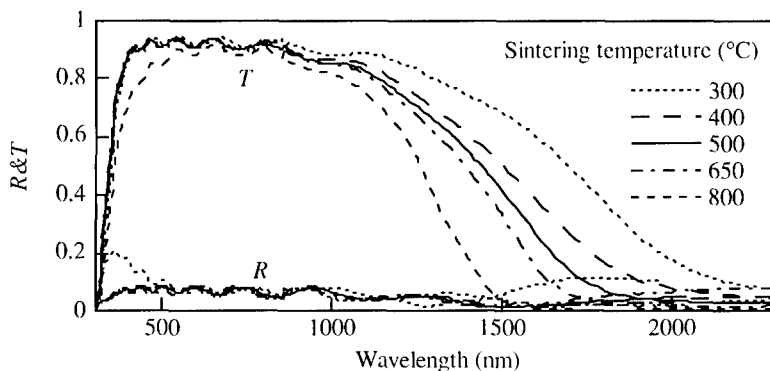
Figure 3: Spectral reflectance (R) and transmittance (T) for nanoparticles-based ITO films sintered at the shown temperatures.
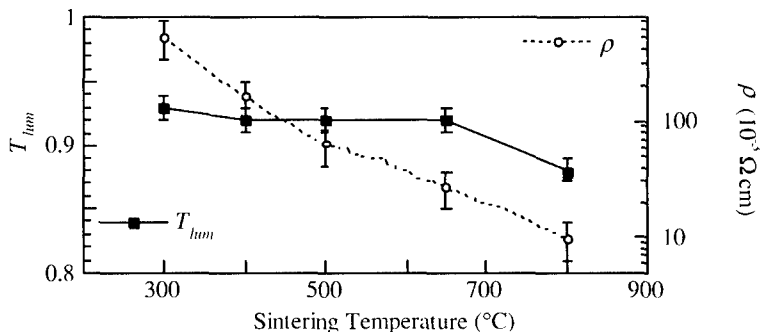

Figure 4: Luminous transmittance (T_{lum}) and resistivity (ρ) as a function of sintering temperature for nanoparticle-based ITO films.

DISCUSSION AND CONCLUSIONS

We made transparent and electrically conducting ITO films by spin-coating a nanoparticle dispersion and ensuing heat treatment. A high luminous transmittance could be combined with a moderately low resistivity, whose value dropped as sintering progressed to higher temperatures. This relationship is connected with grain growth and densification, but no detailed modelling is possible yet. We note that earlier work [6, 7] on sol-gel produced ITO films have stated resistivities as low as 2 to $4 \cdot 10^{-4}$ Ωcm, which are only a factor three off from the lowest resistivity reported for highly transparent ITO layers [8, 9]. A comparison between our data and the former ones is hampered by the fact that the effects of sample ageing were not reported in the cited literature.

ACKNOWLEDGEMENTS

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